Time resolved optical and Raman spectroscopy under ultra-high vacuum conditions: a novel apparatus for pump-probe multi-messenger investigations

C. Fasolato,[1,2] A. Giugni,[3] R. Cucini,[4] A. Fondacaro,[4] G. Panaccione,[4] F. Sacchetti,[1] P. Postorino,[5] C. Petrillo,[1] and G. Rossi[3]

[1] Dipartimento di Fisica, Università degli Studi di Perugia, Italia
[2] Istituto dei Sistemi Complessi – CNR, c/o Sapienza Università di Roma, Italia
[3] Dipartimento di Fisica, Università degli Studi di Milano, Italia
[4] Istituto Officina dei Materiali – CNR, Laboratorio TASC, Area Science Park, Trieste, Italia
[5] Dipartimento di Fisica, Sapienza Università di Roma, Italia
e-mail: claudia.fasolato@cnr.it, andrea.giugni@unimi.it

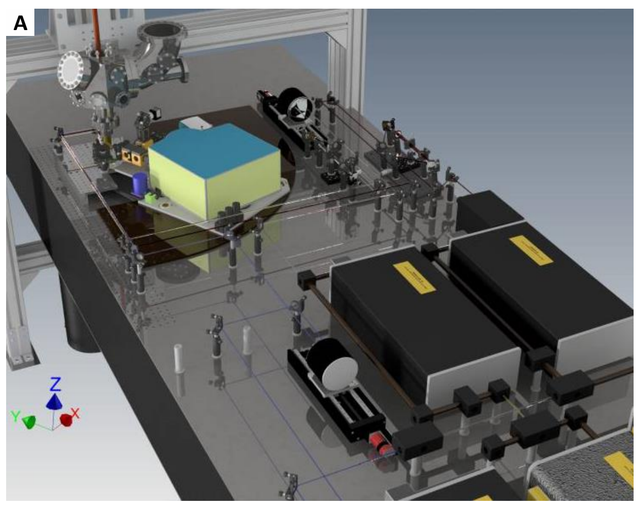
Concept

Our aim is the study of the ultrafast transient dynamics in systems impulsively photoexcited by intense laser pulses. To do so, we propose to synergically combine electronic and optical time-resolved spectroscopies in the pump-probe scheme. To adopt such a multi-messenger approach, the absorbed/scattered photons, photoemitted electrons, and electron-spins are measured as messengers of energy, momentum, and spin of the out-of-equilibrium states after coherent, ultrafast photoexcitation of the system.

At the SPRINT-NFFA facility, we have developed a time-resolved optical and Raman spectrometer for pump-probe experiments at criogenic temperatures and at ultrahigh vacuum conditions (UHV). The chamber is coupled, via UHV-suitcase, with the angle- and spin-resolved photoelectron spectroscopy setups already operative at the facility [1].

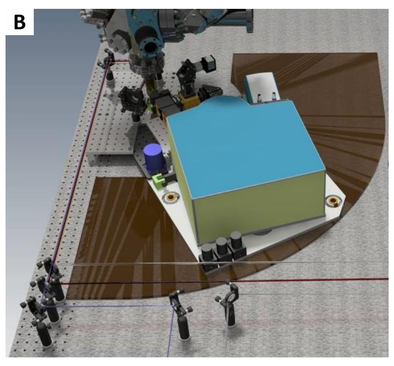
Laser system & excitation

The NFFA-SPRINT laser system, comprises multiple-OPA and a table-top HHG source and can deliver, in a two/three-pulse scheme, ultrashort (~100 fs) IR-to-XUV pump pulses and visible sub-ps probe pulses for Raman and optical measurements.



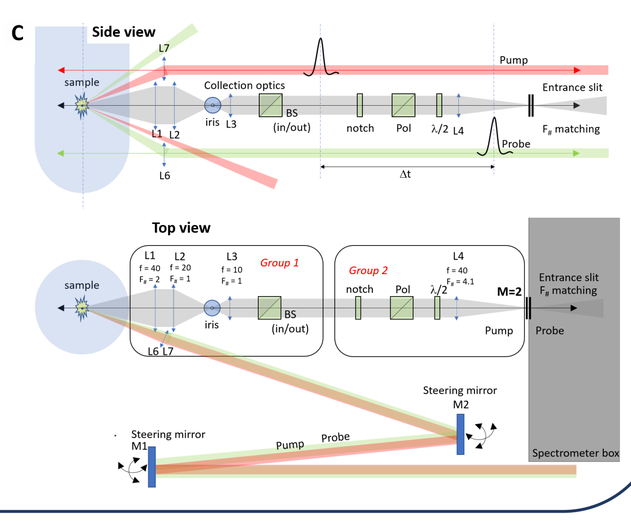
Samples can be excited and probed by the fundamental laser emission (1030 nm), its second harmonics (SH, 515 nm), pulses from the OPAs or a broadband supercontinuum emission, under construction (Fig. A). The relative delay between pulses is controlled by two delay lines, while their spectral shape is controlled by two 4f pulse shapers.

The single pass spectrometer (f = 320 mm) is mounted on a rotating plate, allowing to collect the signal at variable orientation with respect to the sample surface (Fig. B). This allows exploring the scattering selection rules and measuring the optical reflectivity at variable angle, by suitably rotating the sample holder.



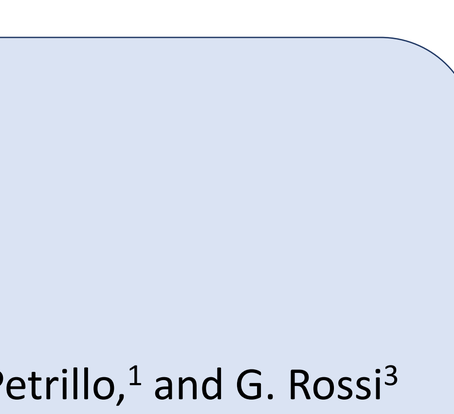
The sample is contained in a sealed off pyrex tube attached to a flange (Fig. C). This allows the beams to access the sample from any direction.

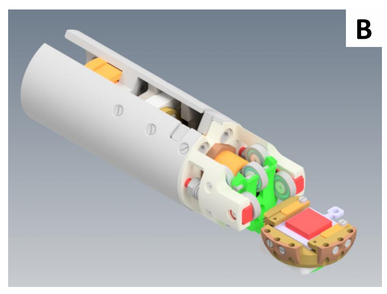
For Raman measurements, high numerical aperture optics is adopted for signal collection. The sample excitation is carried out by separating vertically the optical axes of the pump, probe and collected beams, to minimize the elastic scattering signal and improve the signal to noise ratio.

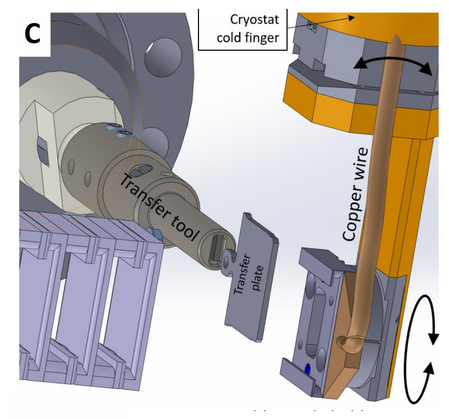


UHV chamber & sample environment

The UHV chamber (Fig. A) features a macroscopic sample manipulator (3 traslations + 1 rotation about vertical axis). The cold finger of the cryostat (He, closed circuit) acts as sample holder featuring two extra rotational degrees of freedom for optimal alignment of sample surface (Fig. B) necessary for reflectivity measurements. The UHV suitcase available at the NFFA-SPRINT facility allows transferring sample between different experimental chambers (Fig. C).

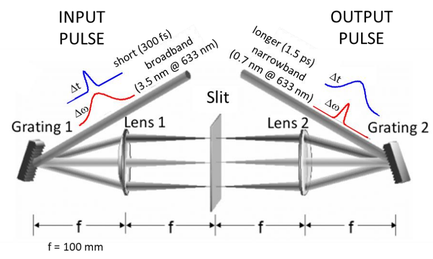




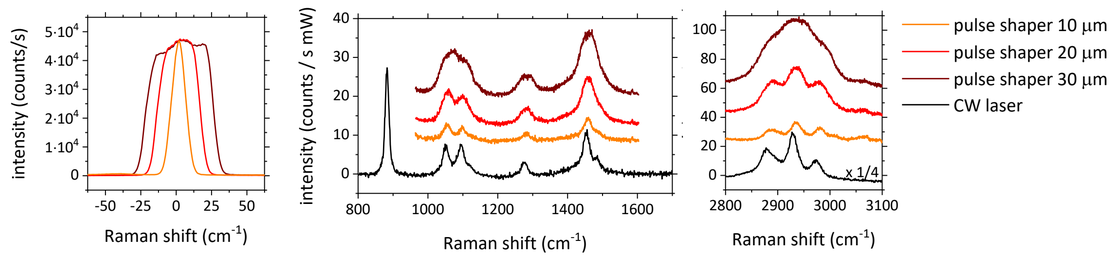


Results of experimental tests

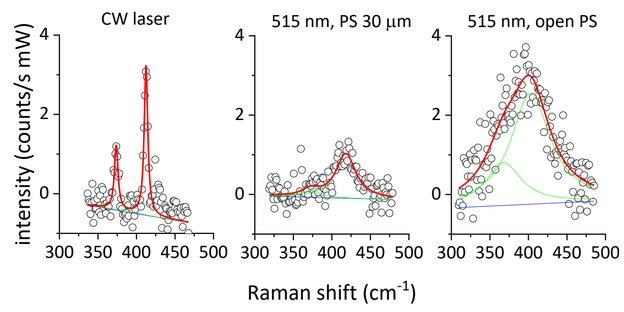
The variable slit opening in the 4f pulse shapers allows to tune the spectral profile of the probe beam. A narrow profile (~15 cm^{-1}) is preferable for achieving highly resolved Raman signals, decreasing the temporal resolution accordingly (~1 ps).



The effect of pulse shaping on OPA 633 nm pulses produces a marked change in the spectral resolution, as visible from the Raman spectra of ethanol, excited by the OPA pulses and compared to a CW laser source.



The same effect can be appreciated in the Raman spectra of a thin sapphire (Al_2O_3) slab, excited by the SH laser pulses at 515 nm.



Perspective experiments

- Hybrid organic-inorganic perovskites ($MaPbBr_3$ and other) as platform for studying the interplay of structure, dimensionality and multi-energy phonon modes concurring to the appearence of **photoinduced transient allotropes and relaxation**. *Time range 100 fs–10 ns*. [2]
- **Coherent light manipulation of magnetic phase transitions** in quantum materials: $MnSe_x$ as a platform for studying selective coherent photoexcitation and collective effects: **variable dimensionality, quantum confinement**. *Time range 50 fs-100 ps*. [3]

References

[1]
[2] H.G. Duan, et al. J. Am. Chem. Soc. 142, 16569 (2020).
[3] D.J. O'Hara et al. Nano Lett. 18, 3125 (2018).

nffa trieste
info.ts@nffa.eu
Q2 Building in AREA Science Park – Basovizza
Strada Statale 14 - km 163,5 - 34149 Trieste, Italy
ph. +39 040 3756487 fax +39 040 226767

ISTITUTO OFFICINA DEI MATERIALI
UNIVERSITÀ DEGLI STUDI DI MILANO
UNIVERSITÀ DEGLI STUDI DI PERUGIA
SAPIENZA UNIVERSITÀ DI ROMA